Canadian Association of Physicists
Association canadienne des physiciens et physiciennes

Contribution ID: **3107** Type: **Invited Speaker / Conférencier(ère) invité(e)**

(I) Plasma assisted oxidation of H_2 mixtures: from physics to chemistry

Wednesday, 8 June 2022 10:45 (30 minutes)

For the past decades, plasma technology has been widely investigated to advance energy and environmental applications. The main focus of this research revolves either around developing new or enhancing existing reforming, combustion, and remediation processes. Nevertheless, due to the complex physicochemical nature of non-thermal plasma's, most of the experimental findings still await a full understanding. Here, we present our recent progress in developing and validating a temperature dependent plasma-chemical kinetic model for H/O/N systems. Our aim is to establish a foundation for low-temperature plasma assisted ignition as well as high-temperature plasma catalysis and reforming processes.

We performed an experimental and numerical study for the plasma assisted low-temperature oxidation of H_2 for undiluted H_2/O_2 mixtures. We employed a temperature controlled dielectric barrier discharge reactor and developed a reaction mechanism as well as a zero-dimensional plasma-chemical kinetic model (KAUSTKin). Through systematically varying the gas temperature and discharge power, we found non-linear oxidation behavior highlighting a Negative Temperature Coefficient (NTC)-like trend in the temperature range of $600 \leq T_g \leq 750$ K. The simulations could successfully attribute these observations to a combination of physical and chemical effects. The unexpected non-linear change in the breakdown voltage and subsequent reduced field (E/N) with varying temperature was the dominant contributor for the nonlinear oxidation characteristics; at the same time, both the O_3 and HO_2 chemistry played a key role.

Our findings show the importance of both the plasma-physical characteristics and the subsequent plasma-chemical kinetics to properly predict plasma processes. Furthermore, the distinct effects of E/N and T_g on the chemistry highlight how electrical discharges provide two ways of controlling the plasma reactions; by affecting the electron induced chemistry (governed by the physical properties of the discharge) and the thermally induced chemistry, respectively. The outcome of this work will serve as a basic building block for future oxidation and reforming studies of CH_4 and higher hydrocarbons.

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Session Classification: W1-7 Non-Thermal Plasmas (DPP) | Plasmas non thermiques (DPP)

Track Classification: Technical Sessions / Sessions techniques: Plasma Physics / Physique des plasmas (DPP)